

Cambridge collaboration probes stratified combustion

Simone Hochgreb, Professor of Experimental Combustion at the University of Cambridge, visited the CRF during July to discuss experiments being conducted in the Turbulent Combustion Laboratory (TCL) by Rob Barlow and Bob Harmon on a turbulent stratified slot burner developed at Cambridge by Ph.D. student Pedro Anselmo Filho. Practical combustion systems are often operated under stratified conditions, either by design in order to promote flame stability while avoiding autoignition, or because perfect mixing cannot be achieved in compact combustors having short residence times. However, stratified flames are not well understood at a fundamental level.

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Laser-induced nanoparticle formation from soot

Sub-micron soot particulates are believed to pose a greater health risk than larger particles and are expected to have a significant impact on the Earth's climate. The growing concern about adverse health and environmental effects of small particles has prompted strict regulations of fine particulate emissions and has intensified research on the formation and impact of combustion-generated particles. Studies of particle formation and evolution, however, are hindered by a lack of sensitive, accurate, noninvasive measurements of their physical characteristics. The

goal of our research program is the development of laser-based diagnostics for *in situ* measurements of volume fraction, size, composition, and morphology of combustion-generated particles with fast time response and high sensitivity. Combustion-generated soot particles consist of branched-chain aggregates of small (15-50 nm diameter) carbon spheroids. In exhaust plumes these particles are often coated with semi-volatile combustion by-products. The nonspherical shapes and inhomogeneous compositions of these aggregate particles make measurement of their physical characteristics difficult, particularly if fast time response and high sensitivity to small particles are required. In addition, laser-induced changes in soot aggregate morphology and fine structure may have an impact on implementation of laser-based detection schemes. Sandia researchers have investigated the physical and chemical changes induced in soot aggregates exposed to laser radiation using results from a scanning mobility particle sizer (SMPS), a transmission

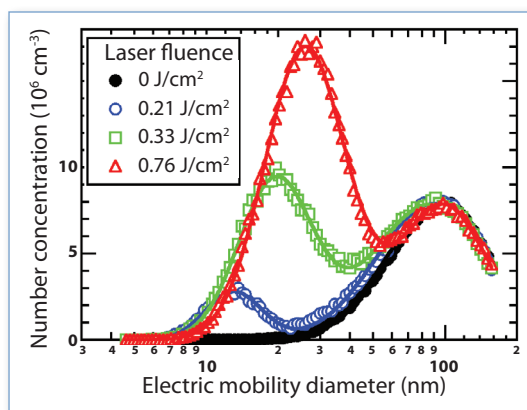


Figure 1. Size distributions of non-irradiated soot particles and soot irradiated at 532 nm with selected fluences.

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New detection system extends the reach of multiscale diagnostics

The use of spontaneous Raman scattering to measure major species concentrations in turbulent flames has a long history at the CRF. Over time, there have been several significant developments in the lasers, detectors, and methods of analysis that have made Raman scattering, in combination with Rayleigh scattering and laser-induced fluorescence (LIF), one of the key experimental tools for fundamental research on turbulent combustion and the establishment of detailed data sets for combustion model validation. Still, the need for simultaneous, single-shot measurements of the state of mixing, the progress of reaction, and gradients in scalar quantities, with both high precision

and good spatial resolution, provides motivation to push these methods toward the physical limits imposed by lasers and optics.

A new detection system for multiscale measurements in flames was recently brought on line in the Turbulent Combustion Laboratory (TCL). As illustrated in Figure 1, this system brings cameras for line imaging of Raman scattering, Rayleigh scattering, and two-photo laser-induced fluorescence of CO into a single unit, which maintains very stable alignment. It combines commercial camera lenses with a custom high-efficiency transmission grating and two motor-driven

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Laser-induced nanoparticle formation from soot (Continued from page 1)

electron microscope (TEM), and a scanning transmission X-ray microscope (STXM) to perform near edge X-ray absorption fine structure (NEXAFS) spectroscopy. These studies were performed by Hope Michelsen and Mark Dansson in collaboration with Mary Gilles and Alexei Tivanski at the Advanced Light Source at LBNL and by Peter Buseck and Laura Van Poppel at Arizona State University.

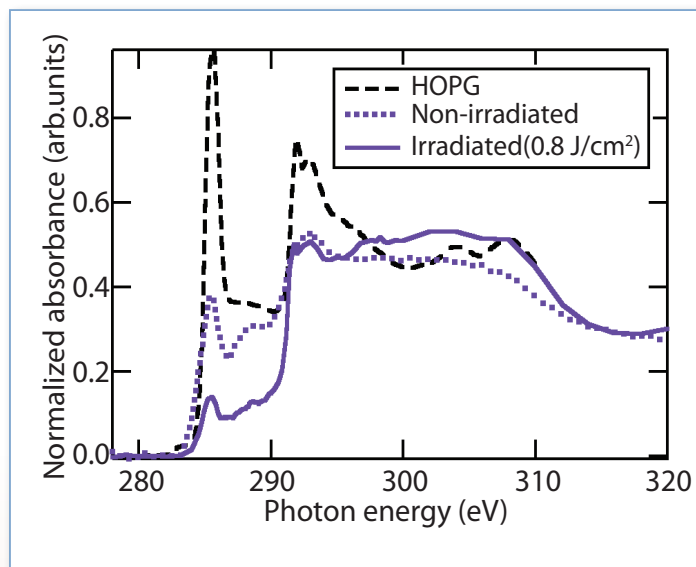


Figure 2. Representative NEXAFS spectra of soot particles before (dotted line) and after (solid line) laser irradiation with a single laser pulse at 532 nm and a laser fluence of 0.8 J/cm². The spectrum of graphite (dashed line) is provided for comparison.

Size distributions of soot particles generated in a flame were measured with SMPS after single-shot laser irradiation at 532 or 1064 nm. Figure 1 shows size distributions of soot particles irradiated with a single laser shot at 532 nm at several laser fluences. The distributions for the laser-irradiated particles demonstrate an additional mode of particle sizes with smaller mobility diameters; this second mode is not apparent in the distribution for the non-irradiated soot (see Figure 1). The mean size and number density of the small particles generated increase with increasing laser fluence.

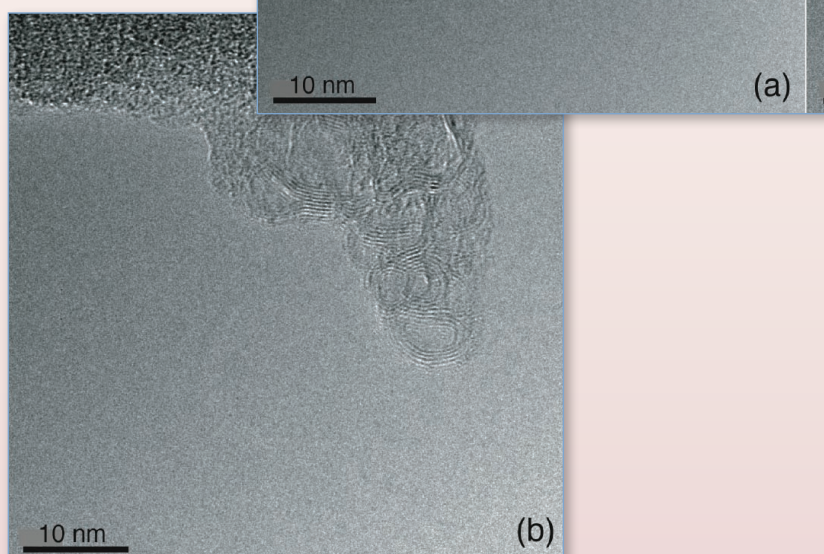
Non-irradiated soot aggregates have a NEXAFS spectrum similar to that of highly oriented pyrolytic graphite (HOPG), as shown in Figure 2. The graphite spectrum exhibits a strong peak at ~285.3 eV, which is attributed to the 1s- π^* transition of aromatic carbon, and a transition at 292 eV, which corresponds to a 1s- σ^* carbon transition with enhancement from a sharp exciton transition at 291.7 eV. The spectrum of the non-irradiated particles also has strong absorptions in these regions. The strength of the 1s- π^* absorption is reduced for particles irradiated with 0.8 J/cm² at 532 nm (Figure 2, solid line), suggesting less graphitic structure.

Spectra from the irradiated particles exhibit a strong exciton at 291.7 eV, however, which could indicate the presence of some short-range order and could be attributable to formation of either annealed structures or amorphous carbon.

Figure 3 shows TEM images of representative nanoparticles produced by laser irradiation at 532 nm at intermediate laser fluences. In many of the particles generated, less long-range order is apparent than in the non-irradiated particles. With 532-nm irradiation at 0.3 J/cm² some of the new particles have no apparent long-range order (Figure 3a), which is consistent with the fine structure of amorphous carbon. Other particles demonstrate structures with layered carbon planes, rings, or folded ribbons (Figure 3b).

Our results show that carbonaceous nanoparticles form during pulsed laser irradiation of soot at 532 and 1064 nm at fluences greater than 0.12 J/cm² and 0.22 J/cm², respectively. Collectively the results suggest that particle growth proceeds through recondensation of small carbon clusters (e.g., C, C₂, and C₃). Nanoparticle growth via homogeneous or heterogeneous nucleation of small carbon clusters is consistent with model predictions of the onset and extent of carbon volatilization by sublimation and photodesorption mechanisms.

Figure 3. TEM images of particles produced at 532 nm with a fluence of 0.3 J/cm² and collected at a mobility diameter of 28 nm.



CRF Hosts Combustion Institute Meeting

On October 16-17, the CRF hosted the Fall 2007 Western States Section meeting of the Combustion Institute at the Martinelli Center, located adjacent to the Sandia site. Over 110 researchers from across the U.S. attended the meeting, which featured 65 technical talks on topics ranging from reaction kinetics to laminar flames to IC engines over the course of a day and a half. Immediately following the meeting, approximately 20 attendees participated in tours of CRF labs. CRF senior manager Andy McIlroy welcomed the attendees and gave a brief overview of the CRF mission and current research areas.

Invited plenary talks were given by CRF's Chuck Mueller and by long-time CRF collaborators Phil Westmoreland of the University of Massachusetts at Amherst and Noel Clemens of the University of Texas at Austin. Chuck discussed the drivers for nontraditional fuel use in high-efficiency, clean-burning engines and provided recent results from his lab on this topic. Phil reviewed the recent advances in flame-sampling molecular beam mass spectrometry and how these advances have revolutionized our understanding of flame chemistry. Noel discussed the challenge of making accurate, laser-based measurements of scalar dissipation in turbulent flames and how important it is to account for spatial resolution and noise effects in a measurement system model.

The conference was organized by CRF's Chris Shaddix and Lyle Pickett, with assistance from CRF office management assistants and the Combustion Institute office.



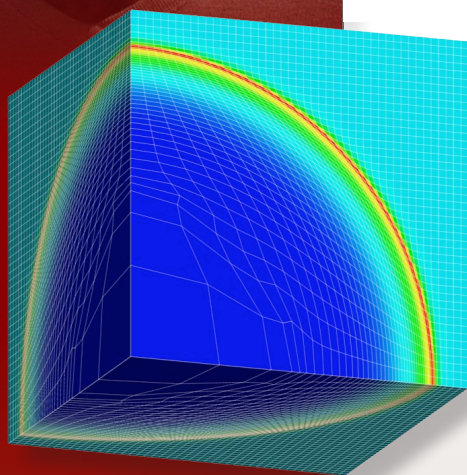
Post-doc and Recent Staff Seminar (PReSS)

Sandia's Post-doc and Recent Staff Seminar (PReSS) series presents monthly hour-long seminars where two post-docs or recently hired staff members talk about their research or other areas of interest. The talks are followed by a question and answer period in an informal atmosphere of a friendly exchange of information.

Variational multi-scale methods in computational mechanics

Guglielmo Scovazzi

Guglielmo Scovazzi at Sandia is studying variational multi-scale methods in computational mechanics. The variational multi-scale framework was introduced in the mid 90's, as an interpretation of stabilization mechanisms in existing numerical methods (particularly, the finite element method). At the same time, it also represents a new pathway for the design of stable, reliable, and accurate algorithms in computational mechanics. The variational multi-scale approach can be interpreted on the basis of purely numerical considerations, as well as from a physics modeling perspective. Applications include shock hydrodynamics, incompressible turbulence, and discontinuous Galerkin methods.



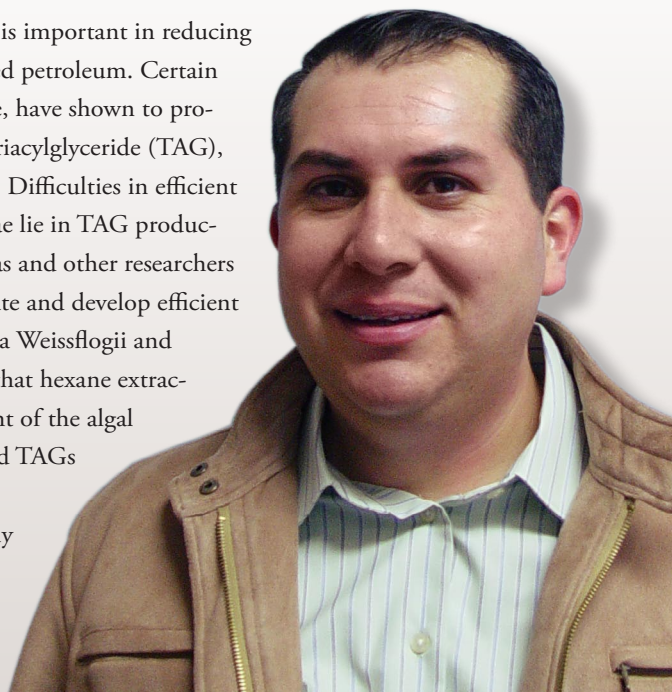
Three-dimensional visualization of a Sedov blast computed on an hexahedral mesh (density contours), a very challenging computation for modern Lagrangian shock hydrodynamics, created by Guglielmo Scovazzi (pictured above)

Extraction and analysis of triacylglycerides from marine microalgae for biodiesel

Frank Zendejas

Production of biomass derived fuels is important in reducing the national dependence on imported petroleum. Certain diatoms, unicellular eukaryotic algae, have shown to produce up to 60% cellular mass, as a triacylglyceride (TAG), used for the production of biodiesel. Difficulties in efficient biodiesel production from microalgae lie in TAG production and conversions. Frank Zendejas and other researchers at Sandia are continuing to investigate and develop efficient techniques for extracting TAGs from *Thalassiosira Weissflogii* and *Thalassiosira Pseudonana*. They have discovered that hexane extractions yield 5 - 10% TAGs based on the dry weight of the algal biomass. The techniques used to analyze extracted TAGs included: Thin-Layer Chromatography for rapid TAG screening process; and Gas Chromatography Mass Spectroscopy techniques to identify the TAG chemical components.

Frank Zendejas pictured

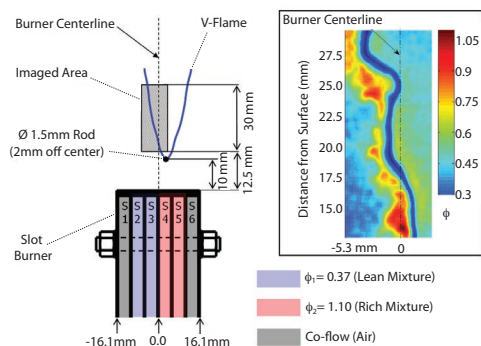


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tion designed for investigation of the fundamental characteristics of turbulent flames propagating through reactants having significant variations in local equivalence ratio. This burner, operating at several stratification ratios and turbulence conditions, has been extensively studied at Cambridge, using simultaneous fluorescence imaging of OH and acetone (seeded into the methane flow), as well as particle image velocimetry (PIV). The example image illustrates the magnitude and scale of the fluctuations in equivalence ratio. Experiments at Cambridge have focused on understanding the effect of stratification on the geometric and kinematic properties of these turbulent flames.

Sandia's contribution was to perform multiscale measurements (temperature and major species), based on simultaneous line imaging of Raman scattering, Rayleigh scattering, and two-photon laser-induced fluorescence (LIF) of CO. These line measurements were combined with crossed planar imaging of OH fluorescence, which yields three-dimensional information on the local flame orientation and curvature in the neighborhood of the line measurements.

The Cambridge slot burner was the first experimental target for a new multiscale detection system in the TCL (see article on p. 1). The combination of good spatial resolution and high collection efficiency in the Raman scattering measurements, which this new system was designed to achieve, is critical for stratified flames because the reaction zones are thin and accurate measurements of the local equivalence ratio are needed.

Figure 2a. Simultaneously measured single-shot profiles (colored symbols) of equivalence ratio, temperature, and the mole fraction of CO across a uniformly premixed turbulent flame. The black lines are from an unstrained premixed flame calculation using CHEMKIN™ with GRI Mech 3.0, with the flame position shifted to match the measured temperature profile.

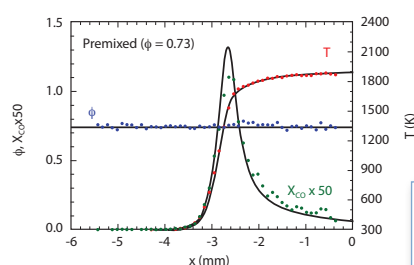


Figure 2a illustrates the performance of this new system by showing measured single-shot profiles of temperature, equivalence ratio, and CO mole fraction, each at 104 micron spacing, in a uniformly premixed turbulent flame ($\phi_1 = \phi_2 = 0.73$). For this realization, the crossed planar images of OH showed a flame front nearly normal to the multiscale measurement line, and the measured profiles are comparable to those from an unstrained premixed laminar flame calculation using CHEMKIN™ and GRI

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Figure 1. In the Cambridge stratified slot burner, one side of a rod-stabilized V-flame intersects a turbulent mixing layer between reactant streams of methane and air having two different equivalence ratios, ϕ_1 and ϕ_2 . The inset (provided by P. Auselmo Filho at Cambridge) shows simultaneous planar images of OH fluorescence from combustion products in the interior of the V-flame and acetone fluorescence from this marker in the cold reactant streams. The color scale corresponds to the equivalence ratio in the room-temperature reactants. The dark blue gap between the acetone (left) and OH (right) sides of the combined image corresponds to intermediate temperatures within the reaction zone.

The slot burner, shown in Figure 1, is a relatively simple configura-

Mech 3.0 (black lines). The steep temperature profile and the peaked CO profile are reasonably well resolved by the experiment, although the CO peak is reduced somewhat by spatial averaging.

The signal-to-noise ratio for equivalence ratio at flame temperatures is between 40 and 50 in these experiments. This precision from the multiscale system is sufficient to measure turbulent fluctuations in the local equivalence ratio in stratified flames, as illustrated in Figure 2b, which includes single-shot profiles from a turbulent stratified flame with stratification ratio 3 ($\phi_1 = 0.37$ and $\phi_2 = 1.10$). The stratification limits are shown as dashed lines, and one can see that there is a significant gradient in ϕ within the thin reaction zone.

The wide range of conditions existing within this turbulent stratified flame is revealed in Figure 3, which compares measured results for CO mole fraction vs. temperature against a family of calculated curves for unstrained premixed flames at several values of ϕ . An interesting feature of this burner is the “tail” of relatively high CO mole fractions that correspond to combustion products at near-stoichiometric or slightly rich conditions.

Analysis of results from a parametric study of flames with various stratification ratios and turbulence levels is in progress, and it is


expected that the combination of diagnostics applied to this burner at Cambridge and Sandia will yield new insights into the structure and propagation of turbulent stratified flames. Further collaboration on stratified combustion in more complex burner geometries is planned, involving both the University of Cambridge and the Technical University of Darmstadt, Germany. 

Figure 2b. Single-shot profiles measured in a turbulent stratified flame with $\phi_1 = 0.37$ and $\phi_2 = 1.10$, as indicated by the dashed lines.

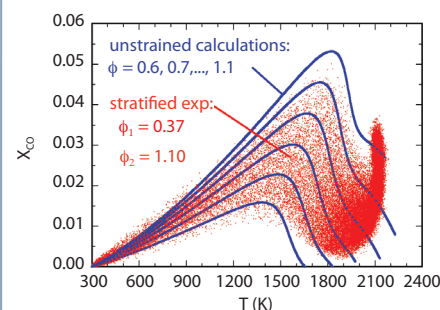
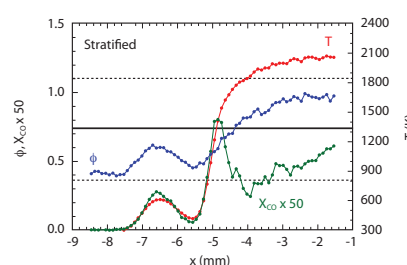


Figure 3. Scatter plot of measured CO mole fraction vs. temperature from a turbulent stratified flame with $\phi_1 = 0.37$ and $\phi_2 = 1.10$. Data were obtained 25 mm downstream of the burner surface, where the average flame position corresponds to the middle of the mixing layer between the two reactant streams. The blue curves are from unstrained laminar premixed flame calculations at the labeled equivalence ratios.

New detection system extends the reach of multiscale diagnostics (Continued from page 1)

shutter wheels that are locked in frequency and phase. The spatial resolution and overall throughput of the Raman system are significantly improved, compared to our previous setup, such that single-shot measurements of temperature and all major species are now obtained with 104 micron spacing of the detection elements (binned pixels). The high-speed (21,000 rpm) shutter wheel provides temporal gating of less than 4 μ s for the Raman measurements, compared to 9.2 μ s with the previous system. This helps the rejection of flame luminosity and allows the use of a non-intensified, low-noise CCD detector with high quantum efficiency.

These improvements are particularly important for measurements of scalar dissipation, which is determined from the instantaneous gradient in mixture fraction and is an important modeled quantity in computational methods for nonpremixed flames. This system

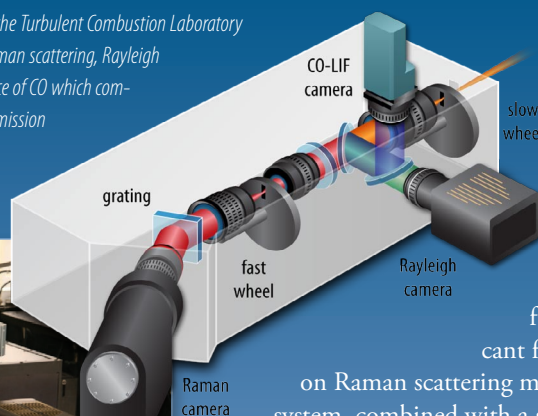
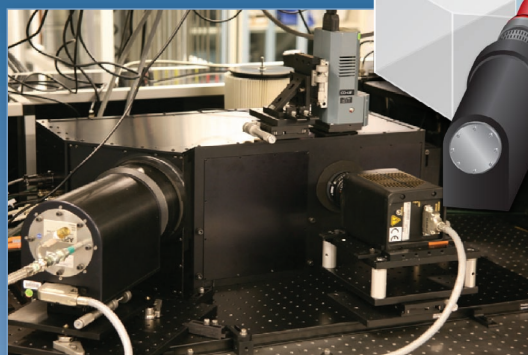
also opens opportunities to extend the application of multiscale diagnostics to a wider range of combustion research problems, including stratified combustion (see article on p. 1), where accurate measurement of the equivalence ratio within thin reactions zones

will give unique insights. Another research area to be explored is the application of multiscale diagnostics to flames of more complex hydrocarbon fuels, including alternative transportation fuels. Flames of complex hydrocarbon fuels can produce significant fluorescence interferences

on Raman scattering measurements, and this new system, combined with a second detector that is under construction, will allow for application of a polarization separation technique to measure and subtract these interferences from the Raman scattering signals. Not all flames will be accessible, but significant extension of these multiscale diagnostics is anticipated.

Several members of the Reacting Flow Research Department and the Engine Combustion Department contributed to the design and development effort, including Guanghua Wang, Rob Barlow, Bob Harmon, Chris Carlen, Duane Sunnarborg, and Uen Do Lee (a visitor from KAIST in South Korea).

Figure 1. The new multiscale detection system in the Turbulent Combustion Laboratory brings together into a single unit, the cameras for Raman scattering, Rayleigh scattering, and two-photon laser-induced fluorescence of CO which combines commercial camera lenses with a custom transmission grating and two phase-locked shutter wheels.



COMBUSTION RESEARCH FACILITY NEWS

Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy under contract DE-AC0494AL85000

CRF News is a bimonthly publication of the Combustion Research Facility, Sandia National Laboratories, Livermore, California, 94551-0969. ISSN 1548-4300

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Editor Frank Cebulski
Graphic Artists Daniel Strong/Michael Takata
Photography Daniel Strong/Various Photographers

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